

MERCURY IN SEDIMENT
AND WATER IN THE
WABIGOON-ENGLISH
RIVER SYSTEM
1970 - 75

JUNE 1976



Ministry
of the
Environment

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MERCURY IN SEDIMENT AND WATER IN THE
WABIGOON-ENGLISH RIVER SYSTEM
1970 - 1975

by
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ONTARIO MINISTRY OF THE ENVIRONMENT
TECHNICAL SUPPORT SECTION
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MERCURY IN WATER AND SEDIMENT OF THE
WABIGOON-ENGLISH RIVER SYSTEM

INTRODUCTION

An estimated 20,000 pounds of mercury were introduced into the Wabigoon River at Dryden, Ontario between 1962 and 1970 by Dryden Chemicals Limited in wastewaters primarily from their chlorine production process which used mercury cells. This has resulted in significant biological changes in the Wabigoon and English River systems and to a lesser extent in the Winnipeg River.^{1,2} In 1970, the Company sharply curtailed mercury losses and, in October 1975, the use of mercury cells ceased. Diaphragm cells were installed and brought into use for chlorine production in December 1975.

Because mercury concentrations in sediment play a key role in methylmercury production, various studies were performed from 1970 to 1975, to ascertain mercury levels in the sediment and whether mercury concentrations were decreasing in the river system. During this period, some studies on water transport of mercury were also conducted. Data used in this report were produced solely by Ministry of the Environment staff with the exception of some data for Clay Lake. An extensive survey of Clay Lake by the Fisheries Research Board of Canada warranted the inclusion of their results.

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

From the water and sediment studies performed on the Wabigoon-English-Winnipeg River system from 1970-75, the following conclusions can be drawn:

- (1) Highly elevated mercury concentrations were found in surface sediments of the Wabigoon River system from Dryden to the outlet of Clay Lake. Some of these surface sediments had mercury concentrations two orders of magnitude higher than the levels found in uncontaminated sediment (approximately 0.1 ppm)
- (2) The highest mercury concentrations were found in sediments near the outfall of Reed Limited at Dryden.
- (3) In the Wabigoon-English-Winnipeg River system below Clay Lake mean mercury concentrations in the top 5 cm. of sediment were less than 0.7 ppm.
- (4) Our data did not show a significant decrease with time in mercury concentrations in sediment in the Wabigoon River from Dryden to Clay Lake during the period 1970-75 or in Clay Lake during the period 1973-75.
- (5) Translocation of large quantities of mercury enriched sediment in the Wabigoon River between Dryden and Clay Lake was not evident during the 1970-75 period.
- (6) Significant amounts of methylmercury were found in Wabigoon River sediment. If sediment conditions remain unchanged, continued high levels of methylmercury can be expected.

- (7) The influence of the physical and chemical properties of sediment on its affinity for mercury and the high spatial variability of mercury levels in the sediments of the Wabigoon-English River system make adequate sampling difficult and hinder efforts to produce truly comparative data over time.

Recommendations

Because fundamental gaps exist in our knowledge of mercury chemistry in sediment and water of the Wabigoon-English-Winnipeg River system, the following studies should be undertaken to elucidate the fate of mercury in this waterway:

- (1) Because of methylmercury's ability to concentrate in the aquatic food chain, information regarding methylmercury concentrations in the waters of the Wabigoon River should be obtained to aid in assessing the long-term ecological effects of mercury presently in the sediment.
- (2) Although sufficient data are now available to give a rough profile of mercury concentrations in Wabigoon River sediments from Dryden to Clay Lake, there has been little sampling immediately below Clay Lake. More intensive sediment sampling should be carried out in the Wabigoon River between Clay and Ball Lakes.
- (3) Because of the nature of certain Wabigoon River sediments, it is expected that mercury-enriched sediment is still being transported down the Wabigoon River

system. Although this quantity may be quite small, it should be measured. In the early 1970's, the Fisheries Research Board of Canada placed a layer of clean inorganic material in Clay Lake to measure the mercury concentration of sediment washed out of the Wabigoon River. Results from this federal study are not yet available, but are expected to be published in 1977. Similar studies should be made downstream of Clay Lake, possibly on Ball and Umfreville Lakes.

MERCURY IN THE AQUATIC ENVIRONMENT

(a) Natural Waters

Natural waters such as the Wabigoon-English-Winnipeg system are highly complex heterogeneous electrolyte solutions, containing organic and inorganic matter in the dissolved, colloidal and suspended states. Besides the ion-water interaction, aqueous solutions are in contact with solids ranging from microscopic to macroscopic dimensions. Thus, other interactions such as ion-ion, ion-solid, and water-solid can play significant roles in the distribution and transportation of mercury between the solid and aqueous phases.

The solubility of mercury under varying water conditions is controlled primarily by the pH of solution, the type and concentration of complexing inorganic and organic ligands and chelating agents, the oxidation state of mineral components and the redox environment of the system.³

(b) Inorganic Chemistry

(i) Eh-pH Diagrams

The classical approach to describing mercury in various oxidation states and compounds present in an aqueous environment is by the use of a Eh-pH diagram. This diagram represents thermodynamically stable species at equilibrium and is not truly representative of natural waters, since natural waters seldom achieve equilibrium and organic compounds are neglected. Organic compounds are not included because they are unstable thermodynamically. The importance of the diagram lies in its ability to illustrate the net

result of complex inorganic interactions and show the different speciation under varying environmental conditions.

Just as pH is a measure of the acidity-basicity of a system, Eh (redox potential) is a measure of the oxidizing-reducing potential of the system. Depending on the redox potential, mercury can be present in three oxidation states which are: (1) elemental (Hg^0), (2) mercurous (Hg^+), (3) mercuric (Hg^{++}).

Oxidizing conditions exhibit positive potentials while reducing environments have negative ones.

In nature, the Eh range is approximately -0.5 to +0.5 volt, and most surface (aerated) waters have a potential of about +0.5 volt. In contrast to the aqueous environment, most sediments are reducing. Even with a constant oxygen exchange at the water sediment interface, reducing conditions prevail a few centimetres below the sediment surface when the organic content is greater than 2% - 5% (John Mothersill - personal communication).

Figure 1 displays the solid and liquid forms of mercury which are stable under environmental conditions in which water itself is stable. From Figure 1 it is apparent that mercury liquid and mercury sulphide will be the prime species affecting mercury solubility in most natural waters.⁴ Figure 2 shows the solubility of various species in equilibrium with the substances shown in Figure 1. Only the main soluble species are shown--those with a mercury concentration greater than 5 ppb. Ammonia, carbonate, bicarbonate and phosphate ions found in natural waters do not affect the relationships

FIGURE 1 - FROM J.D. HEM⁴

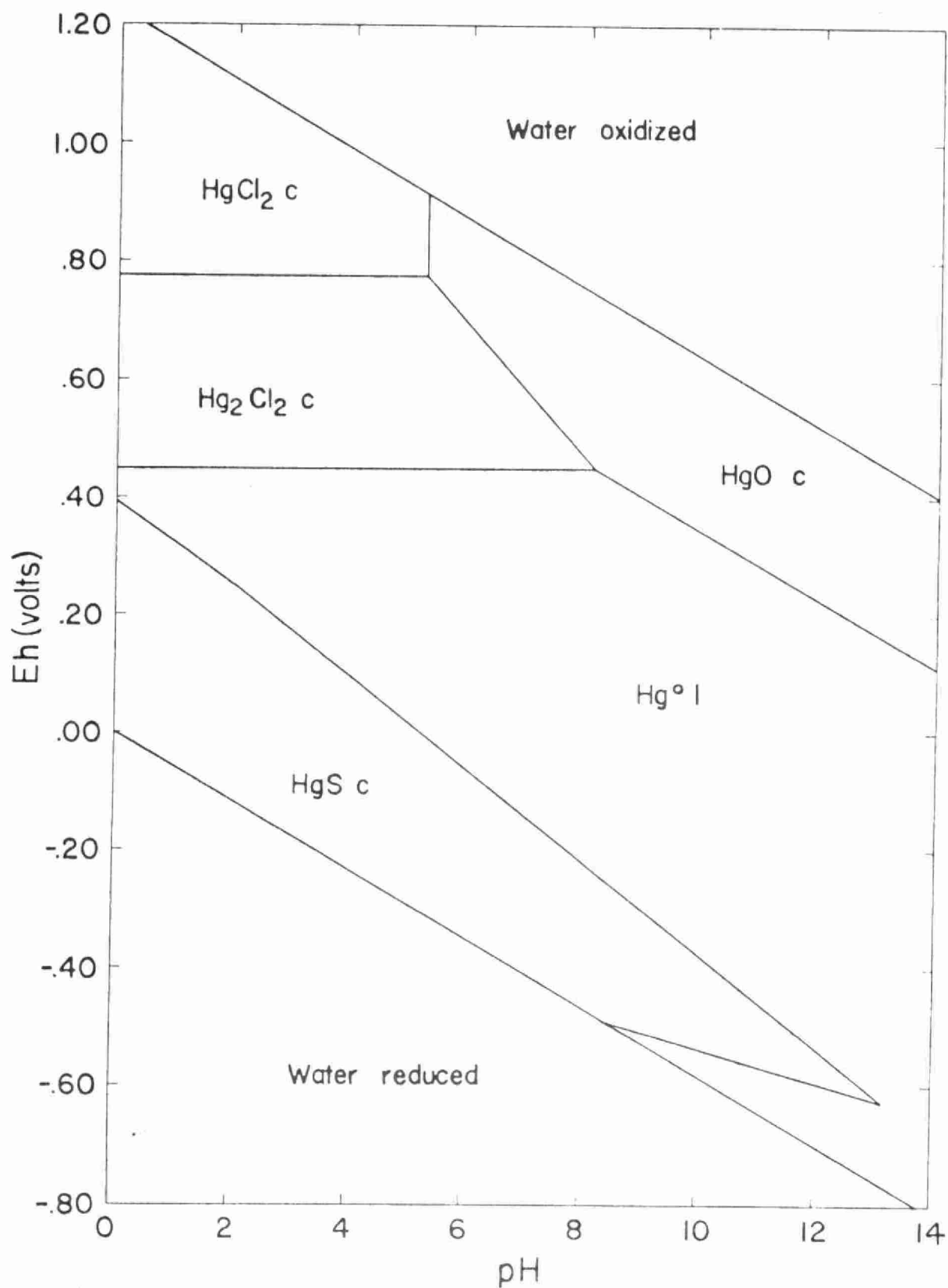


FIGURE 1—Fields of stability for solid (c) and liquid (l) mercury species at 25°C and 1 atmosphere pressure. System includes water containing 36 ppm Cl⁻, total sulfur 96 ppm as SO₄²⁻.

FIGURE 2 - FROM J.D. HEM ⁴

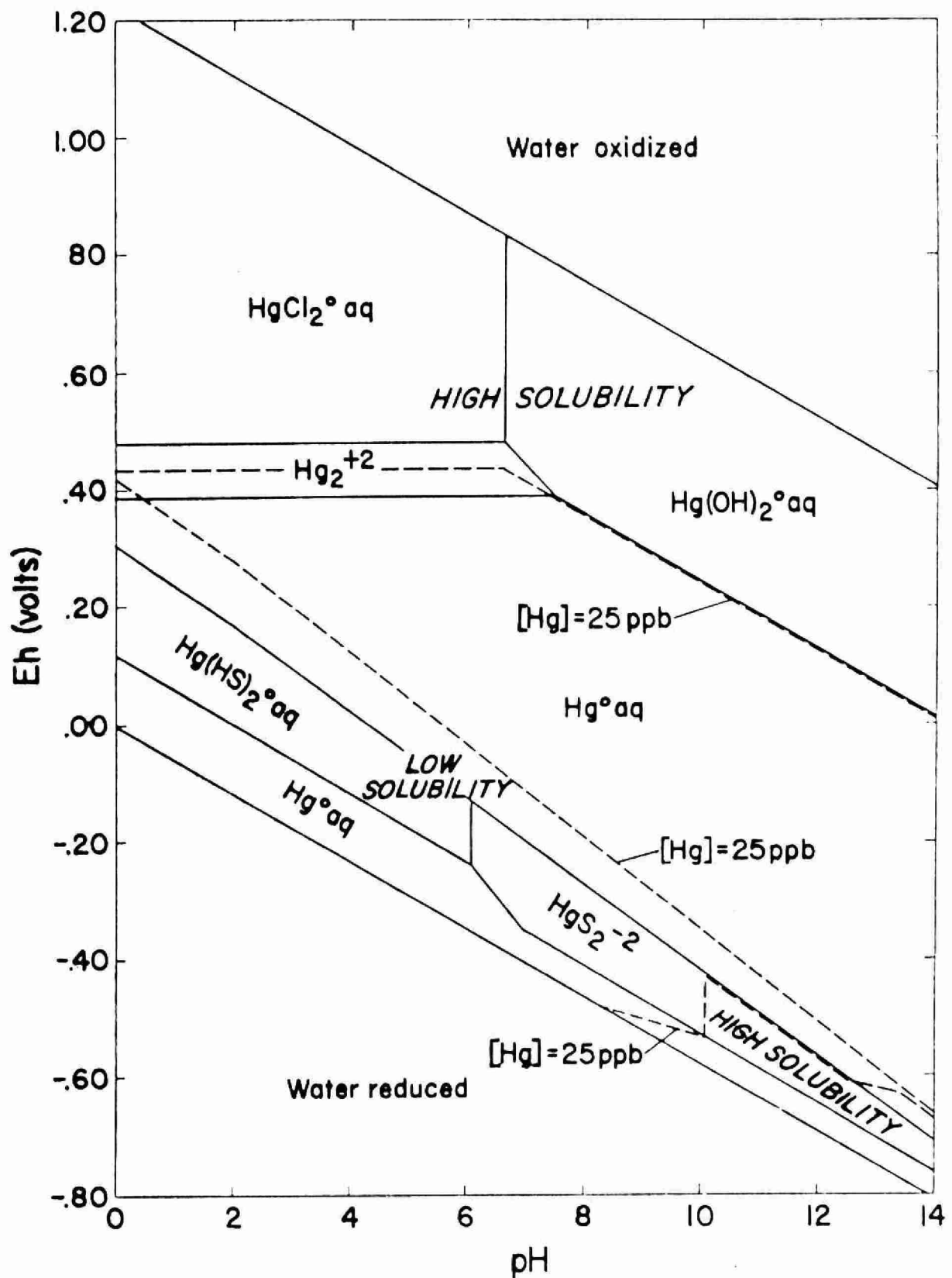


FIGURE 2—Fields of stability for aqueous mercury species at 25°C and 1 atmosphere pressure. System includes water containing 36 ppm Cl^- , total sulfur 96 ppm as sulfate. Dashed line indicates approximate solubility of mercury in this system.

shown by the diagram. The fields of mercury-chloride complexes shrink with lower chloride concentrations, but the mercuric sulphide field stays approximately the same over the range of sulphide ion concentration found in nature. Thus the prominent species in most of the field of Eh-pH values commonly observed is metallic mercury with a solubility of 25 ppb. In well-oxygenated basic waters, the solubility of mercury increases with the formation of $\text{Hg}(\text{OH})_2$ and in chloride-rich acidic waters, increases in solubility are the result of HgCl_2 , and $(\text{HgCl}_4)^{2-}$ complex formation. Since most surface waters have potentials of around +0.5 V, the predominant inorganic soluble species in natural waters will be mercury ions in solution and mercury-chloride, mercury-hydroxide complexes.

Under reducing conditions in the sediment, the presence of sulphide ion effectively immobilizes inorganic mercury regardless of the chemical state of the mercury when it enters the sediment, by formation of HgS .⁵

If the HgS is buried deeply enough, mercury is sufficiently removed from other interactions. However, if the upper layer of sediment becomes oxidized, as is often the case in the spring and fall in mesotrophic and eutrophic lakes, then it is possible for mercuric sulphide to slowly convert to sulphate and the Hg^{++} ion. Since ionic mercury is much easier to methylate than mercury in the form of HgS an increase in the methylation rate of mercury could be expected.

(ii) Adsorption Processes

Adsorption processes of mercury play key roles in the transport and distribution of mercury in fresh waters. Hinkel and Learned, in Gavis and Ferguson⁵, observed that colloidal and particulate matter in natural waters contained 5 to 25 times more mercury than was in solution in the water.

Armstrong and Hamilton, in their work on Clay Lake², found 11% - 75% of mercury in the water column was related to suspended matter. The principal inorganic adsorption processes predominating in the Wabigoon-English-Winnipeg system involve suspended and colloidal clays and silts and possibly iron manganese oxides. These processes are subdivided into two categories--Eh dependent and Eh independent.

Eh Dependent: Inorganic compounds which have solubilities radically affected by Eh changes generated in fresh waters may have some importance in the aquo-Hg cycle. Of principal interest are the iron and manganese oxides which form under a positive redox potential (in aerated waters). With the formation of these insoluble oxides, many heavy metals, including mercury, are co-precipitated.^{5,6} When the redox potential drops and reducing conditions prevail, these oxides may be reconverted to a soluble form, thus releasing mercury back into the water.

Eh Independent: Mercury bonded to silica type compounds is not sensitive to redox changes found in a natural aquatic environment. Clays, silt and sands which are predominantly silica are the key inorganic compounds.

(iii) Particle Size and Mercury Concentration

Among the various properties of sediment, particle size is one of the most significant and other properties (both physical and chemical) are dependent on particle size in a roughly predicatable way. These include shape, specific gravity, mercury adsorption capacity and cation exchange.¹¹ Several investigators^{7,11} have shown that the amount of mercury adsorbed by inorganic material increases with decreasing grain size. It has been found that the capacity of inorganic material for mercury is clays > silt > sand.

(c) Organic Chemistry

(i) Mercury Bonding to Organics

Organic compounds, while usually unstable thermodynamically in a water environment, are very important in the aquatic chemistry of mercury. Divalent mercury has a very high affinity for a large number of organic substances (living and non-living) especially those proteinaceous materials containing the sulfhydryl (R-SH) group. These compounds are stable as long as they are retained by living organisms but upon release by excretion or death of the organism, these compounds decompose and in most cases mercury appears in the inorganic form.⁵ Several studies^{4,7} have shown this high binding capacity of naturally occurring organic sediments for mercury as well as other heavy metals. As soon as organic particles and divalent mercury are present, mercury is very quickly adsorbed on the particulate and eventually settles out.

There is evidence to suggest that certain organic mercury complexes, after sedimenting out, can be reconverted to a soluble form. It has been shown that for mercury humate complexes, the mobility of mercury is very dependent on pH. As the sediment experiences a progressive rise in pH, more and more humate material will become water soluble, taking the bonded mercury with it into solution.⁶ Mercury-humate complexes will play key roles in mercury transport as humic substances are very common, stable, and soluble. Mercury is also known to bond with other complexing agents incorporated in living organisms such as the amino group ($-NH_2$). These complexing groups not only increase the complexing capacity of organics but also increase the strength with which mercury is held.⁵

Organic sulfhydryl complexes exist at redox potentials higher than those at which sulphide sulphur is stable, apparently because of the stability afforded by bonding of the sulphur to the organic residue.⁵ This may have relevance in sediments where the sulphide ion will be removed as a competitive complexing species for Hg^{2+} , leaving mercury available for methylation.

(ii) Methyl and Dimethyl Mercury

The significance of methylmercury was first noted when Westöo found that this form of mercury occurred in fish regardless of whether the source of mercury contamination in the area was phenylmercury, methoxymethylmercury or methylmercury.⁹ Monomethyl mercury (CH_3Hg^+), which acts as a substituted salt and is reasonably water soluble, is found in fresh waters partly as CH_3-Hg^+ ions.

Dimethyl mercury ($\text{CH}_3\text{Hg CH}_3$) remains undissociated in solution and, although it is considered insoluble, there can be enough molecules in water to be of concern. Although this compound is organic, it is not readily decomposed by water. Its stability is probably due to kinetic barriers to decomposition rather than to thermodynamics. In this way the Hg-C bond acts similarly to the C-C bond, which is a remarkably inert although not a particularly strong bond.⁵

The presence of methyl mercury compounds in natural waters was unexpected as there was no readily apparent pathway for their chemical formation. Jernelov⁵, however, showed conclusively that both methyl and dimethyl mercury compounds were synthesized from inorganic mercury in solution by bacteria in the sediment. Since then, several studies have shown that methyl mercury compounds are produced under aerobic or anaerobic conditions. Factors affecting methylation rates and types include pH, organic content, Eh, bacterial populations, temperature and mercury species and concentrations available in the sediment. Acidic conditions favour monomethyl mercury compounds while a basic environment favours dimethyl mercury formation. In the presence of sulphide ion, mercury concentration is too low to allow appreciable utilization by the bacteria. However, upon oxidation of sulphide to sulphate, the mercury is converted to a soluble form and becomes available for methylation. Methylation of mercury from HgS can occur, but the rate is approximately 1,000 times slower than if divalent mercury were present.¹⁰

While it is generally believed bacteria are the prime producers of methyl mercury, fish mucus and certain fungi have also been found to be capable of methylating mercury from its inorganic form.

Besides forming methyl mercury compounds, some bacteria may degrade organomercurials to hydrocarbons and inorganic mercury. This factor complicates estimates of methyl mercury production, since the observed formation of methyl mercury is the net rather than gross methylation rate.⁸

The affinity of CH_3Hg^+ for sulfhydryl groups is about the same as Hg^{2+} although CH_3Hg^+ is able to bond to only one sulfhydryl group, whereas Hg^{2+} can bond to two. Dimethyl mercury itself does not bond to sulfhydryl groups, but at pH below 5.6 it is unstable, as one of the methyl groups will split off and monomethyl mercury will be formed. Once methyl mercury is released into the water it is accumulated and concentrated by living organisms.

(d) Mercury in Sediments

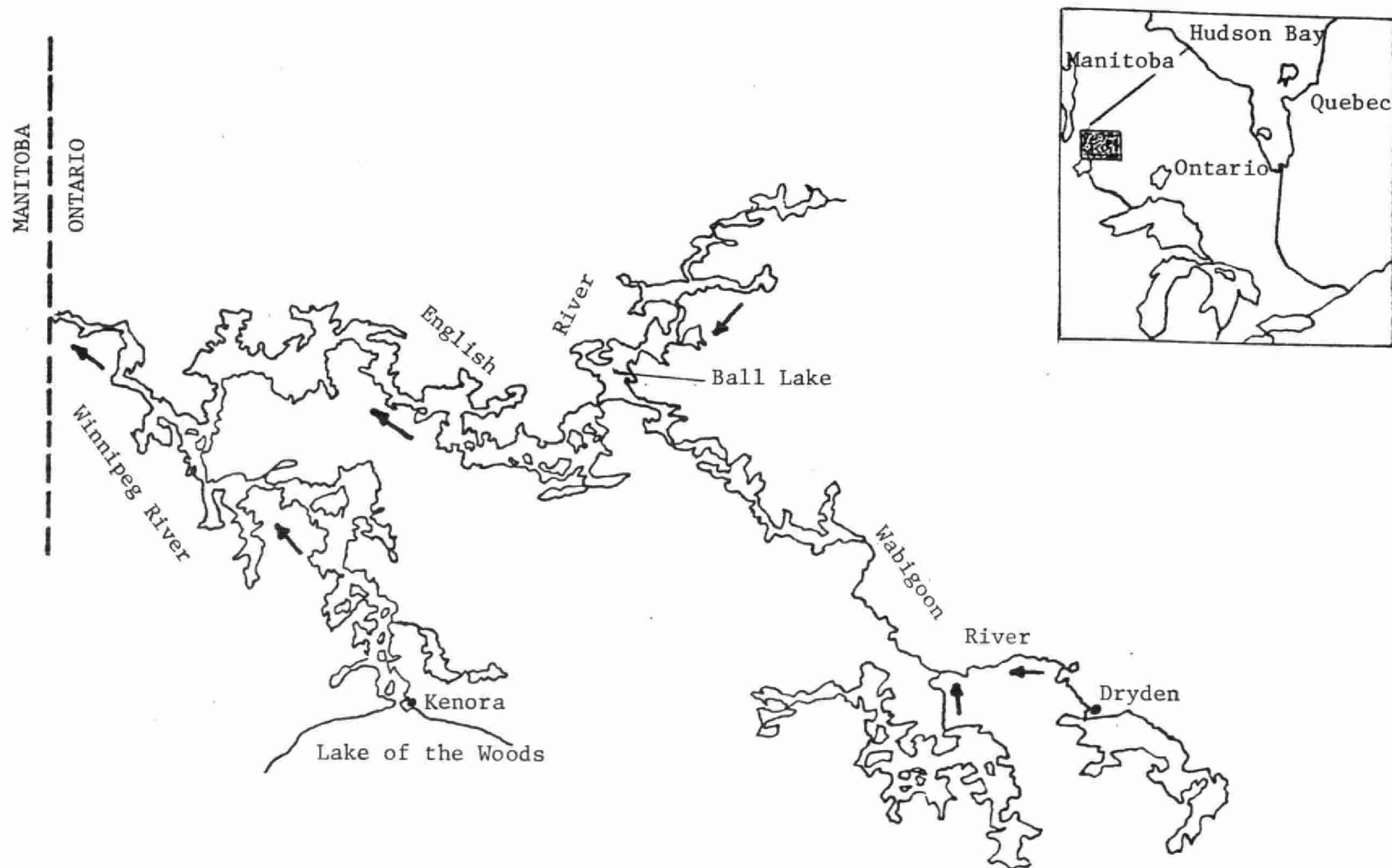
In nature, it is uncommon to find sediments composed of either 100% organic or inorganic material, but rather varying ratios of these sedimental classifications is the norm. A study by Townsend et al¹¹ on sediment having large quantities of wood chips and fibre (not unlike the Wabigoon River) indicates that the highest adsorption capacity of mercury comes from fine particles consisting of clay, silt and fine wood fibres mixed together. Secondary in mercury adsorption capacity were wood chips, followed by the sand

fraction. Reimers et al⁷, came to the conclusion that all forms of particulate organic matter adsorb inorganic mercury, but only sulphur-containing organics have a capacity greater than clay and sands. They also concluded that all organic-rich sediments containing sulphur possessed a high affinity for methylmercury, yet other organic sediment did not appear to have this affinity. The introduction of oxygen into organic sulphide sediments drastically reduced the capacity of these organics for methylmercury, to a magnitude equivalent to natural clays.

Thus, in acidic sediments, mercury levels will be dependent on particle type and size distribution, type and concentration of organic matter and the redox potential.

DESCRIPTION OF STUDY AREA

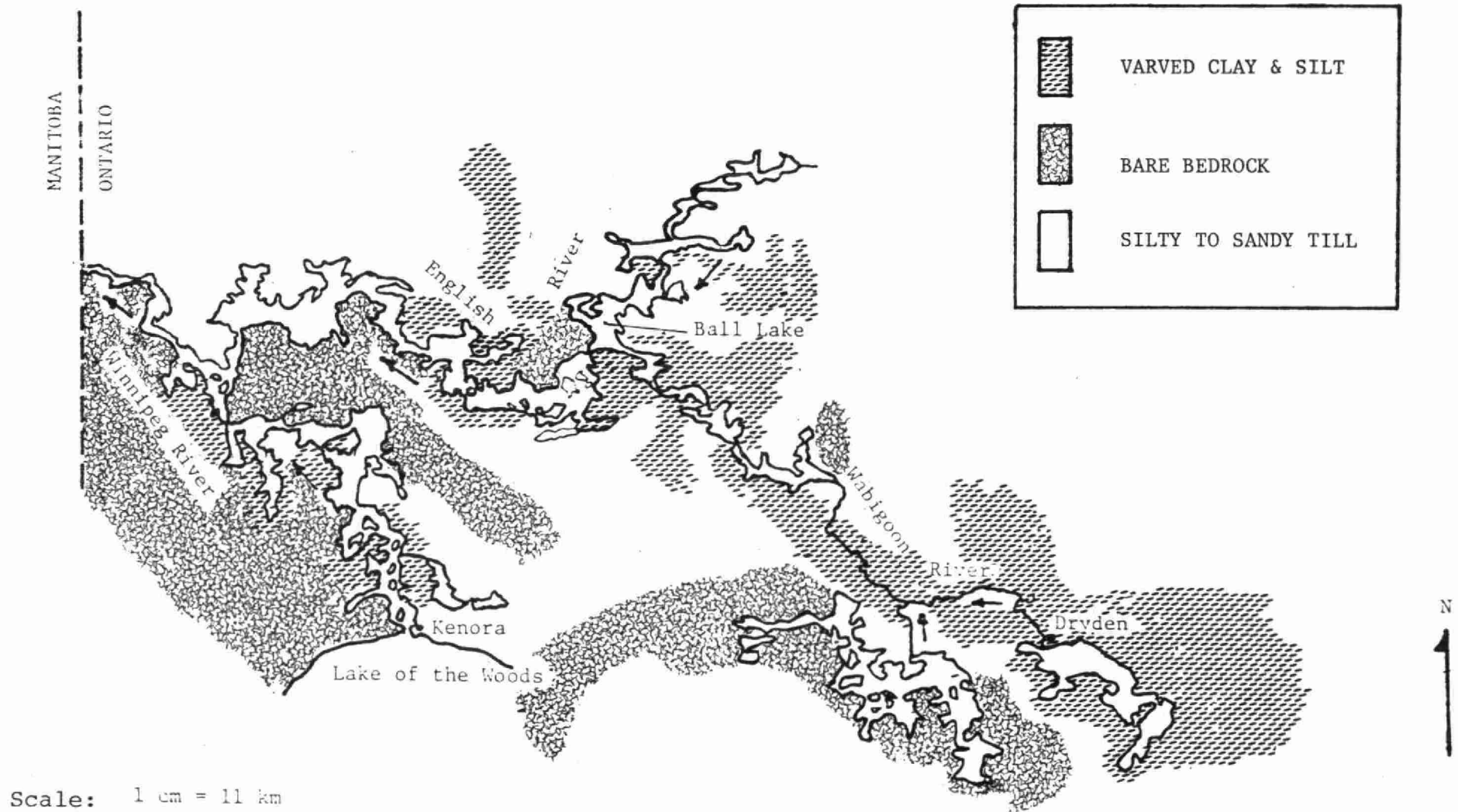
The major region affected by mercury contamination in northwestern Ontario is shown in Fig. 3. Mercury wastes are received by the Wabigoon River at Dryden then flow into the English River at Ball Lake. The English River flows through a series of lakes, eventually joining the Winnipeg River close to the Ontario-Manitoba border. The surficial geology, shown in Fig. 4 indicates that much of the system lies in a region composed of varved clay, silt and sandy till. This explains the high turbidity so characteristic of waters in the Wabigoon-English River system. These mesotrophic lakes have natural sediments consisting predominantly of clay and silt with some sand and organic matter. Increases in organic matter were evident at lake centres.



Scale: 1 cm = 11 km

Figure 3: WABIGOON-ENGLISH-WINNIPEG RIVERS

FIGURE 4: WABIGOON-ENGLISH-WINNIPEG RIVERS
SURFICIAL GEOLOGY



From Ontario Department of Lands and Forests
Kenora-Rainy River Surficial Geology Map S165

SAMPLING METHODS

Due to radical differences in sediment composition, two different methods of sampling substrate were employed. For the most part, in early runs core sampling was used when bottom conditions allowed this procedure. When the river bottom was covered with wood chips, bark and fibre (i.e., the upper Wabigoon River below Dryden), core sampling was impossible and dredges were employed. Table 1 summarizes the sampling method used on the system for the period of investigation.

Because wide variability in mercury concentrations in the Wabigoon River sediments were discovered in 1970, sample intensity was increased by a factor of three for river samples in subsequent yearly runs. Triplicate samples were procured at 1/4 point sections across the stream. To standardize the sampling method, all samples taken in 1973, 1974 and 1975 were obtained by dredge. Since samples taken in 1975 were collected by a different field crew than samples taken in previous years, an additional variable may have been introduced in sampling method.

For studies of water transport of mercury, samples were taken from the water column at 10% width intervals across the river. Subsurface monthly water samples were usually obtained from the river bank.

TABLE 1. Sampling Techniques on Wabigoon-English-Winnipeg River System:

<u>Year and Location</u>	<u>Rivers</u>	<u>Lakes</u>
1970 Wabigoon River to Clay Lake	- for 7 miles downstream of Dryden, 90% of samples were dredged with 3 dredges per site. - below 7 miles all but one sample was cored, with 3 cores per site.	Clay Lake all samples were cored with 3 cores per site.
1971 Clay Lake to Tetu Lake Winnipeg River from Kenora to Tetu Lake	- all samples cored with 3 cores per site.	Samples were cored with 3 cores per site.
1972 Wabigoon River to Clay Lake	- all samples cored with 9 cores per site.	Samples were cored with 9 cores per site.
1973 1974 Wabigoon River, Dryden to Clay Lake	- all samples dredged with 9 dredges per site.	Samples were dredged with 9 dredges per site.
1975 Wabigoon-English- Winnipeg River system Dryden to Tetu Lake	- all samples dredged with 9 dredges per site.	20% with 9 dredges per site. 80% with 3 dredges per site.

RESULTS

WABIGOON RIVER - DRYDEN TO CLAY LAKE

(a) Water and Sediment Characteristics

In studying the pathways of mercury down the Wabigoon, complications arise from the radical change in the aquatic environment caused by the effluent from Reed paper company. During low flows, the Reed paper company discharge (approximately $1.6 \text{ m}^3/\text{sec.}$) comprises 25% of the headwaters of the Wabigoon River. The resulting gross alteration in water and sediment chemistry undoubtedly results in a deviation of the mercury cycle when compared with unpolluted streams, especially when it is realized that many of the components of pulp and paper waste are presently unknown. Severe deterioration in water quality below the mill was evident in comparing upstream data (Table 2) with those from below the mill (Table 3). For stations downstream from the discharge, wide fluctuations in monthly results for some parameters (comparing Tables 4 and 5) were the result of batch discharges from the mill and variations in river flows - the flow in 1973-1974 ranged from $1.4 \text{ m}^3/\text{sec.}$ (51 cfs) to $120.9 \text{ m}^3/\text{sec.}$ (4,270 cfs). The significant increase in suspended solids downstream may have aided mercury transport, due to mercury's strong adsorption characteristics. The effect of heavy organic loadings on dissolved oxygen concentrations is shown in Figure 5. Significant dilution of these contaminated waters is achieved at the confluence of the Eagle River.

An OWRC survey of the Wabigoon River in 1969¹² revealed that "Natural bottom deposits in the Wabigoon River

TABLE 2. Selected water quality data obtained from the Wabigoon River 800 m upstream of mill discharge at Dryden.
Average values for the period 1969-75.

Parameters (ppm)	No. of Samples	Jan.	Feb.	Mar.	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Mean
BOD ₅	55	1.3	1.2	0.8	1.2	1.1	1.2	1.3	1.4	1.5	1.3	1.5	1.2	1.2
TOTAL SOLIDS	45	90	100	100	84	86	130	123	91	89	110	140	67	101
SUSPENDED SOLIDS	54	9	5	3	8	7	20	80	9	8	10	24	3	9
CONDUCTIVITY*	55	102	116	103	100	97	95	94	94	97	127	100	102	101
TURBIDITY **	55	11	11	11	14	12	17	22	18	20	13	25	16	16
TOTAL KJELDAHL	46	.52	.64	.38	.54	.43	.40	.57	.48	.65	.46	.55	.45	.55
CHLORIDES	42			1	2	2		1	2	1	2	2	2	2
pH (Units)	23	7.9	7.6		7.5	7.3			7.7	6.8	7.0			7.4
DISSOLVED OXYGEN	46	12.5	12.3	13.6	13.0	11.7	10.5	9.0	8.2	8.3	11.0	12.2	14.8	11.5

*micromhos/cm

**FTU

TABLE 3. Selected water quality data obtained from the Wabigoon River 1100 m downstream of mill discharge at Dryden.
Average values for the period 1969-75.

Parameters (ppm)	No. of Samples	Jan.	Feb.	Mar.	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Mean
BOD ₅	57	27	41	27	19	9	13	17	14	19	25	20	18	20
TOTAL SOLIDS	47	179	235	220	164	134	265	183	164	214	213	204	150	193
SUSPENDED SOLIDS	58	13	21	16	19	17	16	42	29	28	30	35	25	24
CONDUCTIVITY *	58	221	198	209	164	132	200	191	176	253	239	205	195	191
TURBIDITY **	58	19	20	19	21	22	16	20	27	21	24	25	24	22
TOTAL KJELDAHL	58	.78	.99	.59	.67	.63	.64	.66	.95	2.3	1.9	.72	.69	.89
CHLORIDES	59	25	24	21	9	5	13	11	15	17	25	23	22	17
pH (Units)	28	7.6	7.5		7.4	6.9			7.3	7.3	7	7.7		7.3
DISSOLVED OXYGEN	45	11.3	11.5	11.3	10.2	11.6	8.0		6.0	5.8	7.5	10.7	11.5	9.6

* micromhos/cm

** FTU

TABLE 4. Selected water quality data obtained in 1975 from the Wabigoon River 800 m upstream of mill discharge at Dryden.

Parameters (ppm)	Jan.	Feb.	Mar.	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Mean
BOD ₅	0.8	1.0	0.6	0.8	1.4	0.8		0.8	1.4	1.0	1.0		0.88
SUSPENDED SOLIDS	1	10	1	2	2	10		10	10	5	2		5
CONDUCTIVITY*	99	98	98	100	97	94		91	92	91	94		95
TURBIDITY**	13.0	13.0	13.0	12.0	13.0	19.0		13.0	16.0	13.0	12.0		13
TOTAL KJELDAHL	0.43	0.53	0.34	0.48	0.35	0.28		0.48	0.44	0.44	0.36		0.41
CHLORIDES	1	< 1	< 1	< 1	< 1	< 1		< 1	1	< 1	< 1		< 1
MERCURY (ppb)	0.08		0.1	0.1	.09		.07	.08	.29	.09	.24		.127

*micromhos/cm

**FTU

TABLE 5. Selected water quality data obtained in 1975 from the Wabigoon River 1100 m downstream of mill discharge at Dryden.

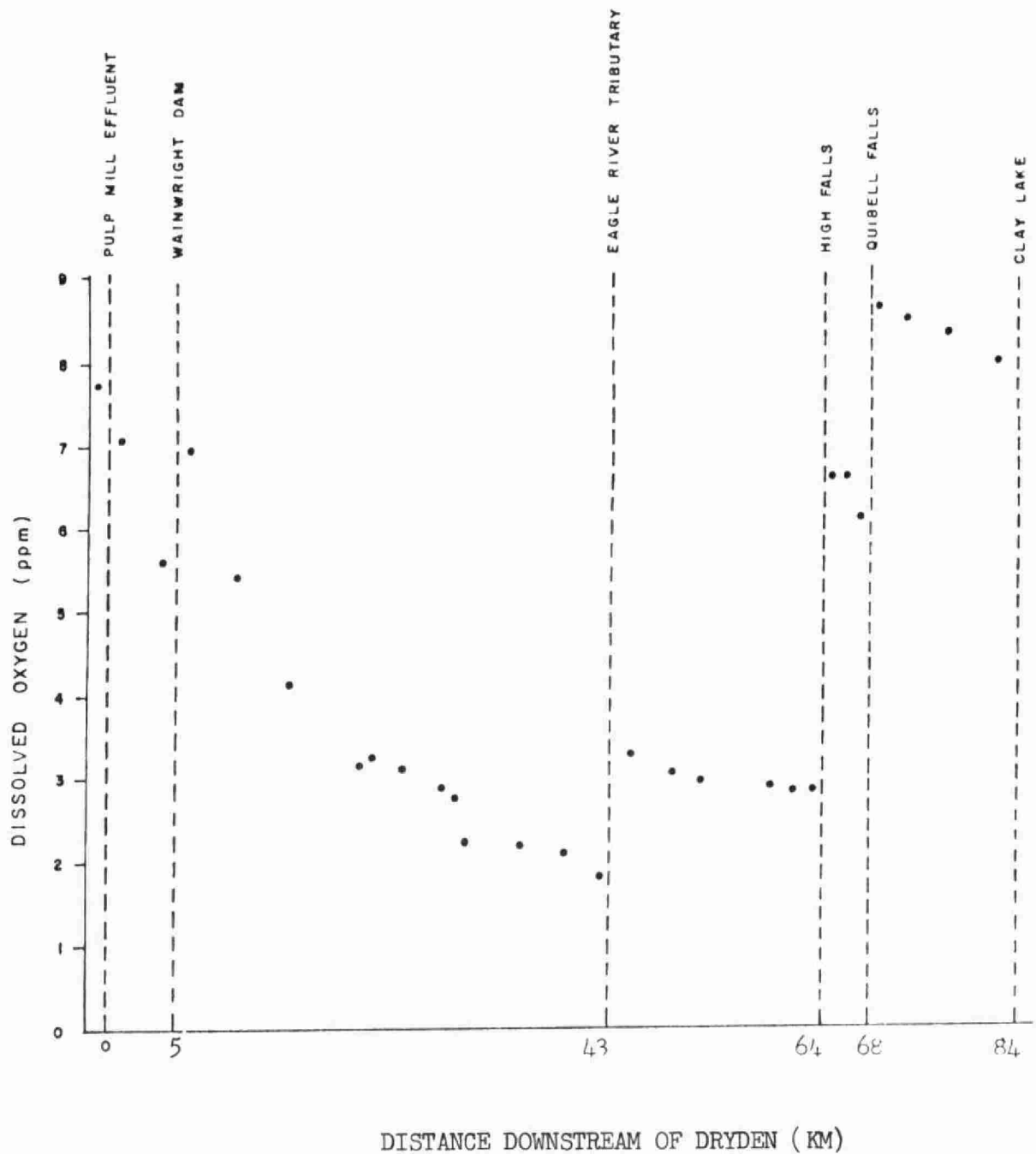
Parameters (ppm)	Jan.	Feb.	Mar.	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Mean
BOD ₅	55	32	22	15	3	30		26	38	28	28		28
SUSPENDED SOLIDS	15	35	20	20	15	30		49	60	35	100		37
CONDUCTIVITY*	320	125	235	132	119	255		190	260	224	146		201
TURBIDITY**	29	33	15	17	26	24		22	23	33	22		24
TOTAL KJELDAHL	.62	.80	.48	.55	.56	.44		.75	.80	.63	.74		.64
CHLORIDES	51	4	27	2	1	27		21	31	26	7		20
MERCURY (ppb)	0.1		15.3	0.1	0.4			0.08	.07	.29	.09	.05	1.8

*micromhos/cm

**FTU

FIG. - 5

DISSOLVED OXYGEN PROFILE OF THE WABIGOON RIVER
SEPTEMBER, 1968 (WATER QUALITY SURVEYS BRANCH) ¹²

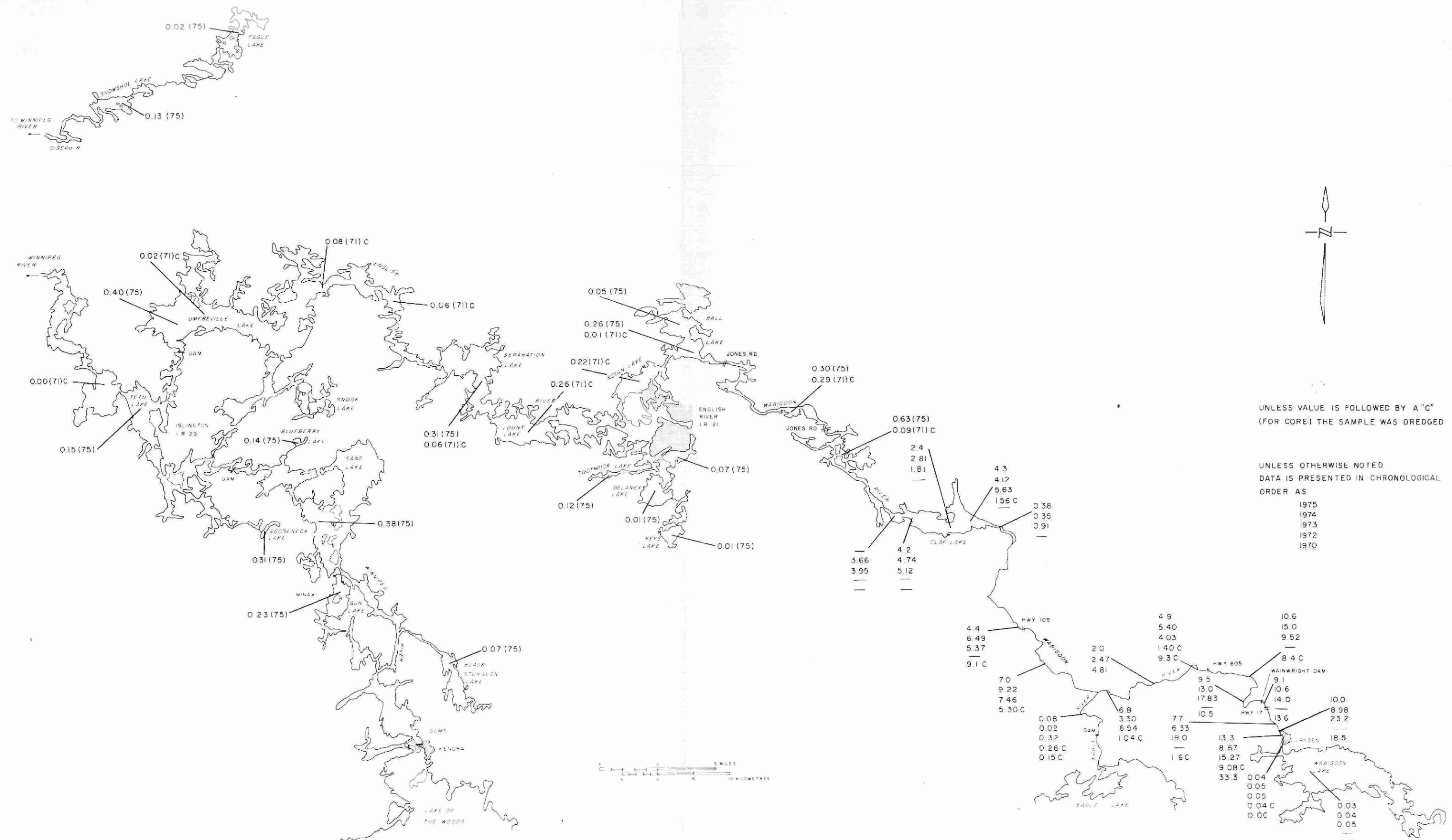


consisted of clay base overlain by soft mud with some isolated patches of sand. Bark accumulations occurred upstream from Dryden from log booming operations. Downstream from the kraft mill to Wainwright Dam (6 km) the natural sediments were completely blanketed by a deep accumulation of bark and coarse wood chips. From Wainwright Dam to Eagle River, a distance of 38 km (24 miles), the substrate consisted primarily of wood fibre, which also occurred in isolated beds as far downstream as Highway 105, a distance of about 64 km (40 miles). Bottom deposits in the Eagle River consisted primarily of sand and clay, much the same as the natural bottom deposits in the Wabigoon River."

(b) Mercury Levels in Sediment and Water

Results for the sampling programs from 1970 to 1975 are shown in Figure 6. Caution must be exercised in interpreting these results as two different sampling techniques were used. Since mercury levels were found to decrease rapidly with increasing depth in sediment, only those values for the top 5 cm were used for comparison purposes. This resulted in modification of some 1971 and 1972 data in an earlier report by J. Bishop in which mercury in sediment values were reported at various depths. Also, 1970 values <0.3 ppm were discarded because their accuracy was in question. Figure 6 indicates that the highest concentrations of mercury were located just downstream of the Reed paper company and mercury concentrations decreased with increasing distance downstream.

FIG. 6: MEAN MERCURY CONCENTRATION IN TOP 5cm OF SEDIMENT (ppm) 1970 - 1975
WABIGOON - ENGLISH - WINNIPEG RIVER SYSTEM



As Bishop noted previously, mercury concentrations in the Wabigoon River sediments were very variable (i.e., it was common for two samples taken at the same location, one metre apart, to have mercury concentration differing by a factor of ten). Further, the sediment was not analysed for either organic content or particle size, both of which significantly influence mercury levels. The absence of data for these important variables made data interpretation very difficult. In Wabigoon River sediments upstream of the mill, mercury levels were 0.1 ppm, but similar levels were encountered downstream only at considerable depth in sediment cores.

The highest mercury concentrations measured (over 100 ppm) were located in the causeway which directly receives mill effluent¹³ (Figure 7). Mill effluent deposits were relatively small and localized. From below the mill to Clay Lake, mercury concentrations in surface sediment were 10 to 100 times greater than upstream values.

In addition to high mercury concentrations, these sediments, composed primarily of wood chips and fibre for 32 km (20 miles) downstream, usually have an oxidizing sediment-water interface. These conditions appear optimum for methylmercury production, values of which are presented in Figure 8. The sediment immediately below the plant contains methylmercury concentrations higher than those found in any sediment sampled in Ontario in which methylmercury can be attributed to microbial methylation. In the river, downstream to Clay Lake, methylmercury concentration was roughly proportional

FIGURE 7: MERCURY CONCENTRATION IN CAUSEWAY SEDIMENT¹³

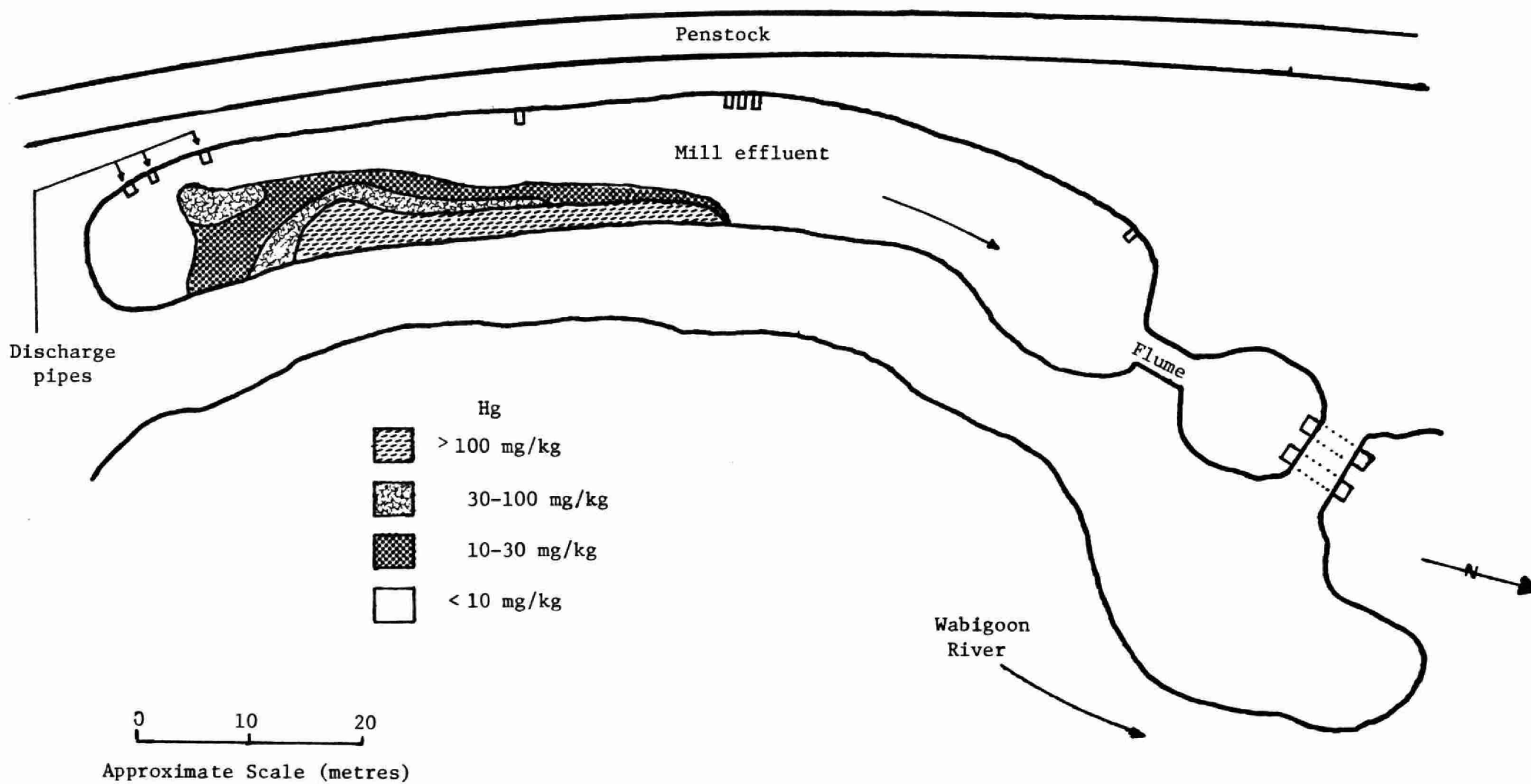
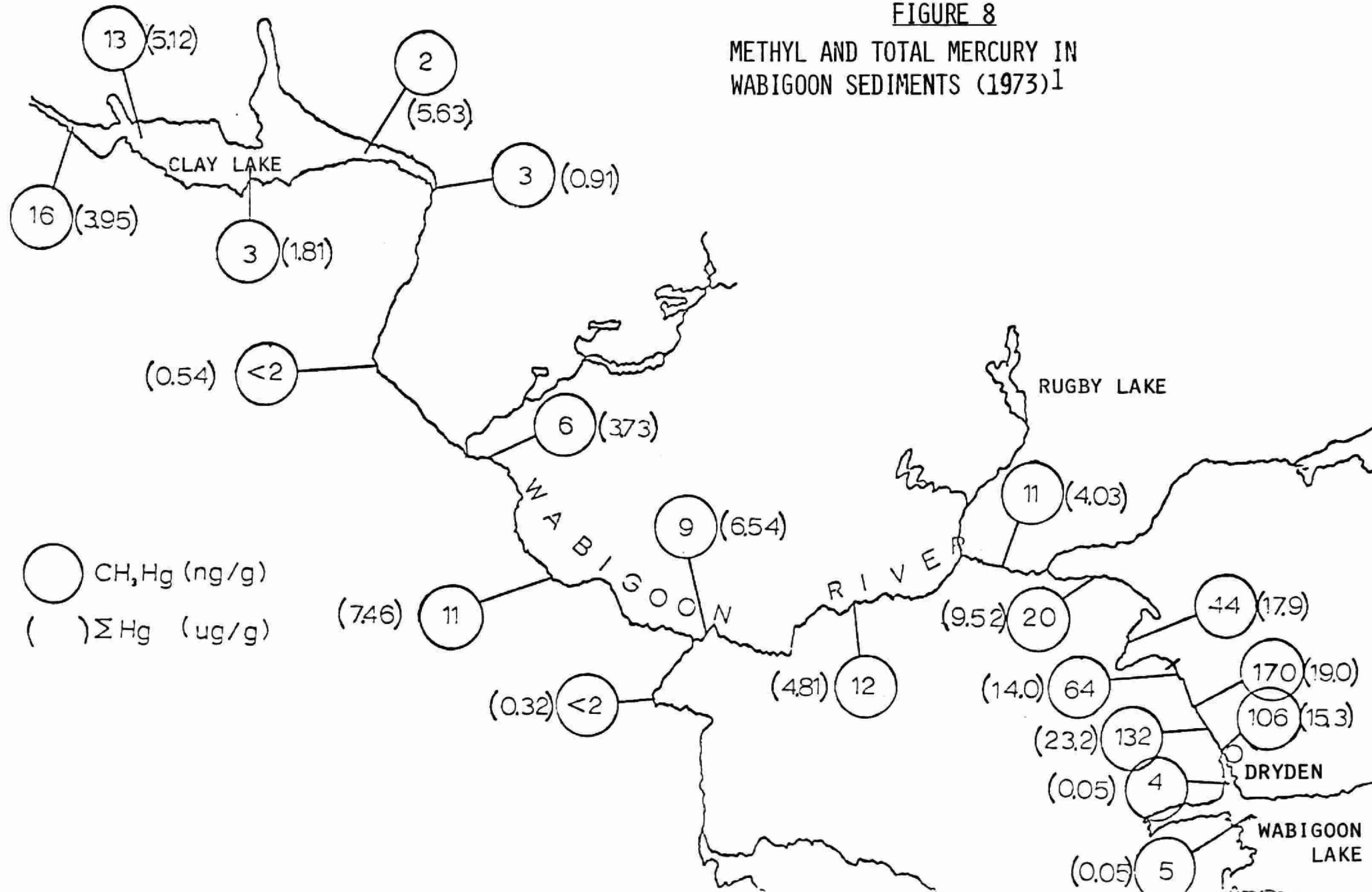


FIGURE 8

METHYL AND TOTAL MERCURY IN
WABIGOON SEDIMENTS (1973)¹



to the total mercury content of the sediment. Therefore, unless there are major changes in sediment conditions, continued high concentrations of methyl mercury are expected to be produced.

The apparent deviation in mercury concentration vs. distance downstream is probably the result of sediment composition. As noted earlier, because of difference in adsorption phenomena a sandy sediment sample taken a few metres from a highly organic one may contain a significantly different mercury concentration. Thus, at any point on the river, mercury concentrations are not only related to distance downstream from the mill but also are dependent upon sediment composition.

Since variables associated with sampling technique and sample substrate were so large, a significant decrease in mercury levels during the 1970-75 period was not evident.

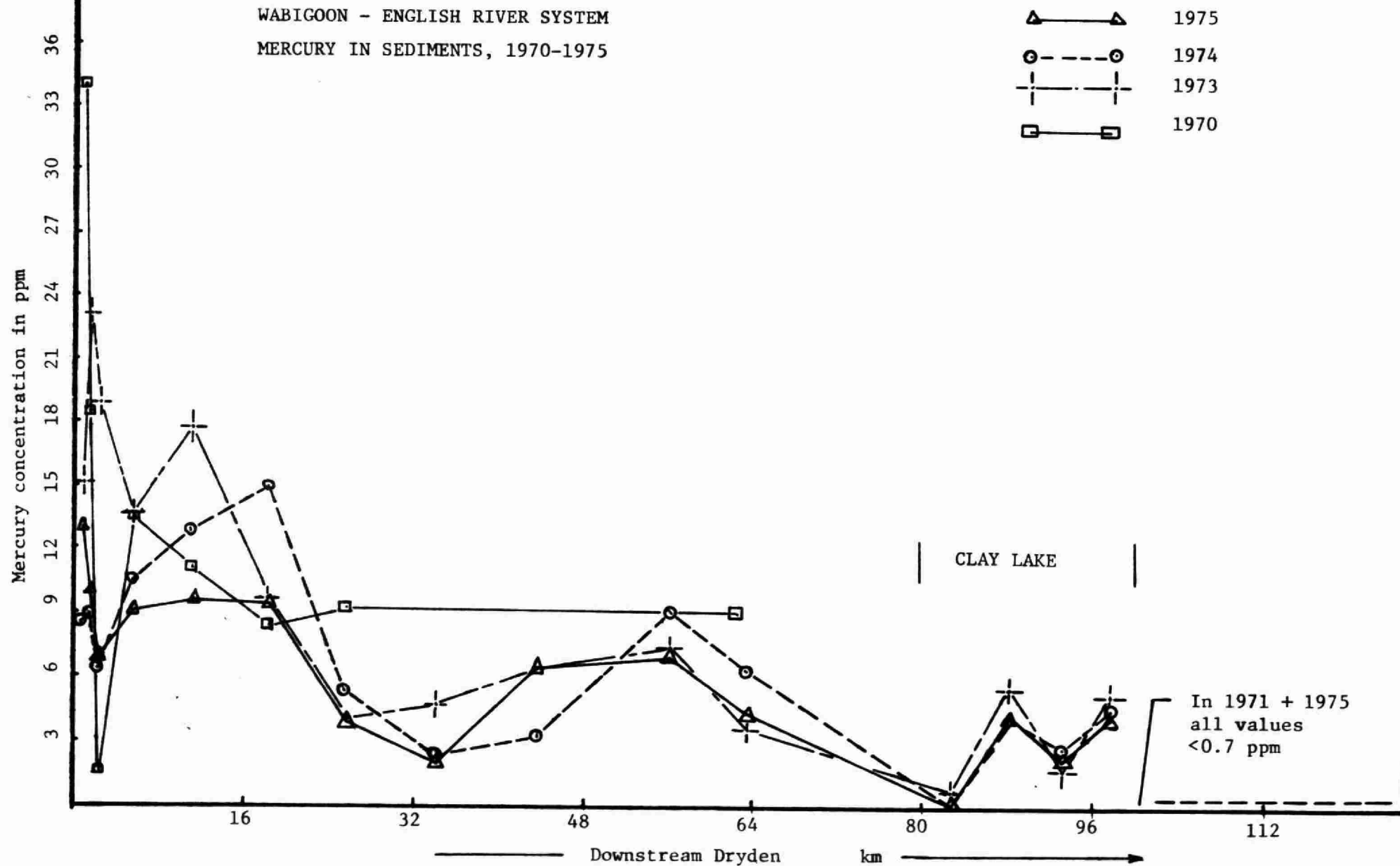
To determine if sediment below the mill exhibiting high mercury concentrations was translocated downstream, levels for five years have been plotted in Figure 9. No radical changes in mercury concentrations were evident during the period in question.

Because water velocity is slowed considerably by the Wainwright Dam 5.5 km (3.5 miles) downstream of the pulp mill, large deposits (up to 12 metres (40 feet) thick) of organic material have accumulated since the dam was built. Although little sampling has been carried out, accumulation of large quantities of mercury in this basin would be in

FIGURE 9

WABIGOON - ENGLISH RIVER SYSTEM

MERCURY IN SEDIMENTS, 1970-1975



agreement with the physical and chemical characteristics of this element.

Core samples of river bottom sediments (Table 6) indicate that most of the mercury is located in the upper layers, especially the top 2 cm. Vertical movement of mercury down the sediment column is not only the result of physical interactions between the suspended material and the receiving sediment, but benthic organisms are also capable of contributing to this action. While certain benthic populations are extremely limited or non-existent on the Wabigoon River sediments because of mill waste, high numbers of pollution tolerant species (sludgeworms) have been observed.

During the period of major mercury losses from Dryden Chemicals Limited (presently Reed Limited), much of the added mercury likely dropped to the sediment as insoluble compounds or accumulated by the settling of suspended sediment enriched by adsorption processes. Upon discharge, mercury entered an aquatic environment relatively high in dissolved and suspended solids. The main chemical competition for any reactive mercury would come from suspended and colloidal organic and inorganic matter as well as complexing agents such as Cl^- , OH^- and S^{--} . Other sulphur compounds identified in kraft mill effluent, like methyl mercaptan (MeSH), could also play important roles in sequestering mercury. A large portion of the mercury presumably adsorbed by the wood fibres and chips, settled out downstream. Mercury which did not settle out remained in the water by bonding on finely suspended and colloidal clays and organics or by forming

TABLE 6. Wabigoon River mercury in sediment data, 1970 (ppm).¹⁴

Sediment Depth (cm)	Distance downstream of Dryden (km)					
	11.2	18.4	25.6	43.2	46.4	63.2
R - 2	9.1	24.0	1.1	9.8	1.1	30.0
4	0.21	19.0	0.09	2.8	0.16	28.0
6	0.03	0.92	0.04	0.35	0.02	16.0
11	0.03	0.38	0.04	0.18	0.04	11.0
21	0.03		0.08	0.10	0.02	0.73
31			0.03	0.07		0.23
41			0.05	0.07		
C - 2	1.4	0.0	5.3	0.15	0.06	0.04
4		0.0	0.88	0.07	0.00	0.30
6		0.0	0.17	0.06	0.02	0.03
11		0.0	0.09	0.09	0.00	0.05
21		0.0	0.05	0.08	0.00	0.04
31		0.0	0.05	0.08		0.03
L - 2	21.0	10.0	24.3	1.1	0.21	0.11
4	35.0	5.9	22.8	0.09	0.12	0.08
6	19.0	1.2	32.1	0.0	0.07	0.08
11	0.16	0.05	2.6	0.0	0.09	0.05
21	0.04	0.03	0.18	0.0	0.03	0.05
31	0.02	0.00	0.08	0.0		0.05
41		0.00	0.09			0.06

R - Right side of river facing upstream.

C - Centre of river.

L - Left side of river facing upstream.

TABLE 7. Wabigoon River - English River - Winnipeg River mercury in sediment data, 1971 (ppm).¹⁴

Sediment Depth (cm)	Distance downstream of Dryden (km)						
	98	125	149	180	213	240	272
R - 2.5	1.6	0.48	<0.01	0.55	0.21	0.07	<0.01
5.0	0.16	0.10		0.36	0.11	<0.01	<0.01
7.5	0.12	<0.01		0.29	0.10	<0.01	<0.01
12.5	0.09	<0.01		0.39	0.07	<0.01	<0.01
17.5	0.08	<0.01		0.31	0.11	<0.01	<0.01
27.5	0.03	<0.01		0.23	0.08	0.04	<0.01
37.5	0.02	<0.01		0.29		0.05	
47.5		<0.01		0.16		0.05	
57.5							
C - 2.5	0.09	0.21	0.03	0.06	0.03	0.00	0.01
5.0	0.07	0.08	0.00	0.04			
7.5	0.06	0.07	0.00	0.04			
12.5	0.06	0.09	0.00	0.04			
17.5	0.05	0.42	0.00	0.04			
27.5	0.00	0.06	0.00	0.03			
37.5	0.02	0.13	0.00	0.04			
47.5	0.02	0.09	0.00	0.04			
57.5		0.07					
L - 2.5	0.12	1.00	0.05	0.31	0.00	0.03	0.00
5.0	0.06	0.33	0.01	0.22	0.00	0.02	
7.5	0.08	0.13	0.00	0.26	0.00	0.00	
12.5	0.04	0.09	0.00	0.17	0.00	0.00	
17.5	0.07	0.07	0.00	0.10		0.00	
27.5	0.07		0.01	0.06		0.00	
37.5	0.07		0.01	0.07		0.00	
47.5	0.06		0.00				
57.5							

R - Right side of river facing upstream.

C - Centre of river.

L - Left side of river facing upstream.

stable complexes ($\text{Hg}(\text{OH})_2$, HgCl_2 , HgCl_4^{--}). Some transport by fine HgS particles would be possible, but eventually this particulate in the water column would be oxidized, releasing mercury. The high pH of the Wabigoon River would favour $\text{Hg}(\text{OH})_2$ formation, but quite likely complexes comprising Hg^{++} , OH^- , and Cl^- exist for some time due to the chloride enrichment of the river from mill wastes. Mercury bonded to suspended and colloidal clays would remain in the water column until conditions (eg. at Clay Lake) favouring flocculation and sedimentation would remove most of it.

Mercury bonded to organics such as fine suspended wood fibres and chips would be released back into the water if these organic compounds were degraded before or after entering Clay Lake. Humic compounds bound to mercury, however, are very soluble and stable and would keep mercury in solution. Methyl mercury compounds diffused from the sediment would be very stable and would be dispersed down the river system.

Mercury transport studies conducted since 1970 on the Wabigoon during high and low flows reveal low quantities of mercury in the water column. In 1970, water samples collected on the Wabigoon River from Dryden to the outlet of Clay Lake had < 10 ppb mercury. Sampling in 1972 was conducted at the Golf Course Bridge--approximately 1.6 km (1 mile) downstream of Dryden Chemicals--and at Highway 605--approximately 25.6 km (16 miles) downstream from Dryden Chemicals--during low flows. Of the 53 water samples collected, 45 contained less than 0.10 ppb mercury and four contained

trace levels (0.15 ppb). Three samples at Highway 605 contained 0.16, 0.35 and 0.44 ppb mercury and one contained 190 ppb at the Golf Course Bridge. During the spring run-off in May, 1975, 20 samples were taken at the Golf Course Bridge when the mill was shut down. Nineteen samples had a mean of 0.08 ppb (range 0.06-0.10) and one result was 0.6 ppb. Unfortunately, comparisons cannot be made between these data and values obtained above the mill because of wide variations (.01-.29 ppb) in upstream values. Intensive sampling of Lake Wabigoon and the Wabigoon River above the mill and chemical plant in 1976 should result in more useful data for comparison purposes.

Water samples taken monthly in 1975 at the Golf Course Bridge (Table 4) also revealed wide variations in mercury concentration. Varying river flows and batch discharges from the mill caused significant fluctuations in many water quality parameters. The anomalous value of 15.3 ppb (Table 4) could be the result of sampling error or the entrapment of sediment particles forced into the water by gas formation in the sediment. Because of the variation in mercury concentrations a study is now being undertaken to assess whether the variation is real or the result of sampling error.

CLAY LAKE

Due to morphological differences in lakes and rivers, their sediment, water and mercury cycles differ significantly. Like many lakes in Northwestern Ontario, Clay Lake has lakeshore sediments composed primarily of silt and fine sand with some larger grained materials. Silt,

and organic matter are more abundant towards the lake centre. Maximum mercury accumulation would be expected in sediments near the centre of the lake because of their probable greater ion exchange capacity than that of sediments at other locations. Also, optimum conditions for flocculation and sedimentation of colloidal clays would be encountered at the lake centre.

Although much of the mercury waste discharged at the Dryden mill was bonded to river sediment, a significant portion has also been deposited in Clay Lake. Armstrong and Hamilton² estimated this quantity to be approximately 2,000 kilograms. They also found that over 90% of the mercury above background concentration of 0.1 ppm was in the top six centimetres of sediment. Mercury concentrations varied from 0.1 ppm at shorelines to 7.8 ppm in the deeper waters. The average value for samples analysed was 3.1 ppm. They noted that although the highest concentrations of mercury were elongated in the direction of water movement, considerable mercury deposition had occurred in the northern arm of Clay Lake. They also found that the highest mercury concentrations occurred in the eastern basin of Clay Lake, although the western basin is considerably deeper (24 metres vs. 6 metres) and has slower water movement.

Although Ministry of the Environment data for Clay Lake are limited, the available data generally agree with Armstrong and Hamilton's results. Analysis of our data revealed no significant change in mercury concentration in Clay Lake during the period 1973 to 1975.

WABIGOON-ENGLISH-WINNIPEG RIVER SYSTEM
CLAY LAKE TO MANITOBA BORDER

Limited sampling has indicated substantially lower mean mercury levels (less than 0.7 ppm) in surface sediment downstream of Clay Lake to the Manitoba border. From cores taken (Table 7) it is evident that surface sediments were enriched with mercury in comparison to deeper substrate. As mentioned previously, for comparison purposes the data was standardized to the top 5 cm. However, it should be noted that the top 2.5 cm. of sediment have mercury values higher than those results shown in Figure 6. Comparison of data from different years would be misleading, as samples taken in 1971-75 were collected using different sampling techniques and locations and insufficient data are available.

OFF SYSTEM LAKES

The results of the top five cm. of dredged sediment from off system lakes (for location see Fig. 6) is presented below:

<u>Lake</u>	<u>Mercury Concentration (ppm)</u>
Keys	.01
Toothpick	.12
Delaney	.10
Snowshoe	.13
Eagle	.02
Blueberry	.14
Sand	.38
Gooseneck	.31
Black Sturgeon	.08
Gun	.20

Unfortunately these lakes were not core sampled and comparison of upper and lower sections of cores to determine if recent mercury enrichment took place was not possible. Seven of the ten lakes, however, have mercury concentrations of approximately 0.1 ppm which is in agreement with uncontaminated sediment of the Wabigoon-English River system.

ACKNOWLEDGEMENT

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APPENDIX - Mercury in Sediment of Wabigoon-
English-Winnipeg River System 1970-1975

Facing Upstream

L = Left side of River

C = Centre

R = Right side of River

WABIGOON RIVER MERCURY SEDIMENT DATA 1970 (ppm)

Sediment Depth	Above Source				Below Source		(km)					
	-7	-5	1	15	2.5	6	11	18	26	63		
0-2cm	0.00	0.00	Dryden Chemicals Ltd.	19.	32.	3.9	15.	9.1	24.	1.1	30.	
2-4		0.00						.21	19.	.091	28.	
4-6		0.00						.032	.92	.044	16.	
6-11	R	0.00						.027	.38	.040	11.	
11-21								.034		.081	.73	
21-31										.034	.23	
31-41										.047		
0-2cm	0.00	0.00		25.	21.	.029	4.9	1.4	0.00	5.3	.044	
2-4		0.00				.023			0.00	.88	.30	
4-6	M	0.00				.29			0.00	.17	.031	
6-11		0.00				.099			0.00	.086	.053	
11-21		0.00							0.00	.048	.038	
21-31									0.00	.050	.034	
0-2cm	0.11	0.00		56.	5.4	1.7	21.	21.	10.	24.3	.11	
2-4	.071	0.00			.91	.21		35.	5.9	22.8	.077	
4-6	.065	0.00			.22	.080		19.	1.2	32.1	.077	
6-11	.072	< .01			.064	.090		.16	.05	2.6	.051	
11-21		< .01			.037	.084		.038	.028	.18	.054	
21-31								.024	.00	.081	.054	
31-41									.00	.091	.060	

LEGEND: R - Right side of river facing upstream
 L - Left side of river facing upstream
 M - Midstream

Single values represent a dredged sample.

WABIGOON RIVER-ENGLISH RIVER-WINNIPEG RIVER SYSTEM MERCURY IN SEDIMENT DATA, 1971 (ppm).

Sediment Depth (cm)	Distance Downstream of Dryden (km)										
	98	113	125	149	157	180	192	213	224	240	272
R - 2.5	1.6	.42	0.48	< 0.01	.36	0.55	.09	0.21	.08	0.07	< 0.01
5.0	0.16	.00	0.10		.15	0.36	.00	0.11	.01	< 0.01	< 0.01
7.5	0.12	.00	< 0.01		.07	0.29	.00	0.10	.00	< 0.01	< 0.01
12.5	0.09	.00	< 0.01		.06	0.39	.00	0.07	< .01	< 0.01	< 0.01
17.5	0.08	.00	< 0.01		.09	0.31	.00	0.11	.01	< 0.01	< 0.01
27.5	0.03	.00	< 0.01		.06	0.23	.00	0.08	.01	0.04	< 0.01
37.5	0.02	.00	< 0.01		.04	0.29	.00			0.05	
47.5		.00	< 0.01			0.16				0.05	
C - 2.5	0.09	.03	0.21	0.03	.24	0.06	.02	0.03	.32	0.00	0.01
5.0	0.07	.05	0.08	0.00	.14	0.04	.00		.05		
7.5	0.06	.02	0.07	0.00	.29	0.04	.03		.04		
12.5	0.06	.02	0.09	0.00	.14	0.04	.02		.03		
17.5	0.05	.03	0.42	0.00	.34	0.04	.02		.02		
27.5	0.00	.03	0.06	0.00	.00	0.03	.01		.02		
37.5	0.02	.02	0.13	0.00	.00	0.04	.02		.02		
47.5	0.02	.02	0.09	0.00		0.04			.04		
57.5		.01	0.07								
L - 2.5	0.12	.01	.99	0.05	.34	0.31	.23	0.00	.01	0.03	0.00
5.0	0.06	.01	0.33	0.01	.12	0.22	.02	0.00	.01	0.02	
7.5	0.08	.00	0.13	0.00	.09	0.26	.21	0.00	.01	0.00	
12.5	0.04	.00	0.09	0.00	.08	0.17	.11	0.00	< .01	0.00	
17.5	0.07	.00	0.07	0.00	.12	0.10	.11			0.00	
27.5	0.07	.00		0.01	.11	0.06	.13			0.00	
37.5	0.07	.00		0.01	.14	0.07	.16			0.00	
47.5	0.06	.00		0.00	.11		.14				
57.5											

R - Right side of river facing upstream
C - Centre of river
L - Left side of river facing upstream

WABIGOON RIVER MERCURY IN SEDIMENT DATA 1972 (ppm)

Sediment Depth		Distance from Dryden Chemicals Ltd. (Km)						
1 = 0-2.5 cm		-.5	+1	26	43	55	88	Eagle R.
2 = 2.5-5 cm								
L	A-1	.010	15.84	0.95	1.64	.238	2.367	0.018
	A-2	.028	11.08	0.06	.137	.047	0.056	.014
	B-1	.018	5.25	4.08	.344	.448	0.119	.021
	B-2	.031	10.43	0.40	.017	0.038	.040	.021
	C-1	.015	11.30	3.03	.420	.665	3.662	.023
	C-2	.023	7.10	1.83	.020	.029	0.650	.024
C	A-1	.050	10.49	.088	.397	8.84	5.000	.008
	A-2	.026	---	0.05	.141	2.02	.241	.023
	B-1	.016	11.13	4.34	.302	4.69	4.434	.016
	B-2	.022	---	0.15	.028	1.53	.220	.006
	C-1	.019	5.33	0.08	1.52	9.73	1.976	.015
	C-2	.053	3.98	0.03	.110	2.81	.061	---
R	A-1	.050	8.94	4.0	1.20	6.98	.068	.055
	A-2	.130	26.61	0.198	0.243	.087	.060	.050
	B-1	.016	7.66	5.31	2.84	4.86	3.790	.038
	B-2	.064	8.08	0.065	2.56	.080	.217	.020
	C-1	.019	4.26	0.516	5.61	22.56	4.230	.014
	C-2	.049	5.85	0.023	1.26	22.82	.842	.013
		.04	9.1	1.40	1.04	5.3	1.56	

WABIGOON RIVER SYSTEM - MERCURY IN SEDIMENT 1973 (ppm)

<u>Distance Downstream of Dryden Chemicals Ltd.</u>	<u>Top 5 cm of Dredge</u>		
(km)	L	C	R
-7	0.04	0.04	0.06
-.5	0.04	0.04	0.08
1	20.5	19.6	5.70
1.5	23.2	23.0	23.4
2.5	26.7	15.5	14.8
6	12.8	12.8	16.5
11	20.7	12.0	20.8
18	20.6	3.49	4.44
26	4.39	3.52	4.19
34	2.95	5.64	5.83
43	8.47	7.19	3.90
55	10.6	5.20	6.31
63	1.50	4.88	9.74
83	0.94	0.83	0.95
88	5.87	4.43	6.60
91	0.19	4.93	.30
98	3.78	6.07	5.52
101	5.81	1.81	4.22
Eagle River	0.35	0.34	0.27

WABIGOON RIVER SYSTEM - MERCURY IN SEDIMENT 1974 (ppm)

<u>Distance Downstream of Dryden Chemicals Ltd.</u>	<u>Top 5 cm of Dredge</u>		
(km)	L	C	R
-.5	13.3	3.83	9.82
+1	6.0	6.0	14.1
1.5	13.3	3.83	9.82
2.5	6.32	4.97	7.69
6	12.8	10.56	8.42
11	15.5	10.4	13.0
18	15.41	16.4	13.1
26	7.05	8.02	1.14
34	1.58	1.62	4.20
43	3.48	0.93	5.50
55	16.2	3.57	7.88
63	0.18	8.19	11.1
83	0.35	0.52	0.18
88	4.06	3.07	5.22
91	0.22	4.63	3.59
98	3.46	5.55	5.21
101	5.34	5.08	0.56
Eagle River	.03	< 0.01	0.03

WABIGOON-ENGLISH-WINNIPEG RIVER SYSTEM - MERCURY IN

SEDIMENT 1975 (ppm)

Distance downstream
of Dryden Chemicals Ltd.

Top 5 cm of Dredge

(km)

L

C

R

-7	.01	.05	.04
- .05	.02	.04	.06
1	29.0	2.3	8.5
1.5	0.63	11.4	18.0
2.5	4.8	11.1	7.1
6	9.6	9.4	8.4
11	9.0	9.0	10.6
18	12.0	8.8	11.0
26	5.0	6.1	3.6
34	2.0	3.0	1.0
43	5.2	7.4	7.8
55	7.4	3.8	9.7
63	0.56	---	8.2
83	0.27	0.57	0.32
88	3.2	5.4	4.2
91	2.3	0.41	4.4
98	5.0	3.8	3.8
113		.63	
125		.30	
149	0.05	0.24	0.28
170	0.06	0.04	0.1
192	0.40	0.32	0.22
246		0.40	
264		.16	
		.14	

Clay Lake

Clay Lake

Clay Lake

Clay Lake

Ball Lake

Grassy Narrows L.

Separation Lake

Umferville

Tetu Lake

Off System Lakes

Keys Lake	0.01
Toothpick	0.12
Delaney	0.10
Snowshoe	0.13
Eagle	0.02
Blueberry	0.14
Sand	0.38
Gooseneck	0.31
Black Sturgeon	0.08
Gun	0.20



(7743)

TD/427/M4/P37/MOE

[illegible]